Band tail absorption saturation in $CdWO_4$ with 100-fs laser pulses

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In scintillators based on exciton luminescence, the phenomenon of nonproportionality can be explained in terms of nonlinear interactions between relaxed exciton states [1-4]. For well-known scintillators like $CaWO_4$, $PbWO_4$ and $CdWO_4$, whose luminescence originates from self-trapped excitons, the Förster energy transfer mechanism of dipole-dipole interaction (FRET) leads to the nonlinear losses of luminescence for excitons created in particle tracks. In order to find the most important parameter for quantifying nonproportionality, the Förster interaction radius, optical studies at high excitation densities have so far been performed with excitation energies that fall into the fundamental absorption region, where the unknown values of the absorption coefficient restrict an accurate determination of the Förster radius. In order to avoid such uncertainty, the present study was performed using sub-band-gap excitation energies (the Urbach tail region), were the absorption coefficient is low enough to be directly measured. However, this requires taking into account an additional effect that is not evident in the fundamental absorption region.

For sufficiently high excitation densities excitonic absorption saturation in $CdWO_4$ gives rise to induced transparency [5]. Phonon-assisted absorption, generally the accepted mechanism of absorption in the Urbach tail, cannot be applied to fs pulses, i.e. when pulse duration is shorter or comparable to the period of optical phonons. Instead, absorption is determined by the availability of suitable atomic displacements from equilibrium positions such that the difference in excited and ground state energies equals the excitation energy at the moment of excitation. The number of centers with an atomic configuration favorable for excitation is limited, which is the reason for saturation. A model combining saturated absorption and FRET is presented, which allows to study the saturation effect by analyzing the decay kinetics of excitonic luminescence. In $CdWO_4$ the Förster radius is calculated to be 3.7 nm. Maximum exciton densities in $CdWO_4$ (with a band gap of 5 eV) are calculated as 5.3×10^{18} cm^{-3} for 4.1 eV photons and 1.9×10^{19} cm^{-3} for 4.66 eV photons. The approach developed is expected to be useful for similar experiments using powerful lasers pulses.

References

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